Lightning and anthropogenic NOx sources over the United States and the western North Atlantic Ocean: Impact on OLR and Radiative Effects

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Abstract

The migration of enhancements in NO_2 concentration, outgoing longwave radiation (OLR), and radiative effects associated with the onset of the North American Monsoon in July 2005 has been investigated using satellite data and the Regional chEmical trAnsport Model (REAM). The satellite data include the tropospheric NO_2 columns, tropospheric O_3 profiles, and OLR from OMI, TES and NOAA-16 satellite, respectively, for June and July 2005. The simulated OLR captures the spatial distribution of the remotely sensed OLR fields with relatively small biases (≤5.7%) and high spatial correlations (R≥0.88). This study reveals that the lightning-generated NOx exerts a larger, by up to a factor of three, impact on OLR (up to 0.35 Wm^{-2}) and radiative effects (up to 0.55 Wm^{-2}) by enhancing O_3 in the upper troposphere than anthropogenic NOx that increases O_3 in the lower troposphere, despite the fact that the lightning-generated NOx and O_3 are much smaller than those from the anthropogenic emissions. The radiative effect by lightning-derived upper tropospheric O_3 over the convective outflow regions is affected by the changes in lightning frequency. Thus the changes in convection due to global warming may alter the geographical distribution and magnitude of the radiative effect of lightning-derived O_3, and this paper is a first step in quantifying the current radiative impact.

1. Introduction

Nitrogen oxides ($NOx=NO+NO_2$) are major ozone (O_3) precursors produced primarily from fossil fuel combustion, lightning, and soil. Among these, NOx from anthropogenic emissions and lightning are the most important sources of O_3 in the lower and upper troposphere, respectively, over North America and the North Atlantic Ocean [Ridley et al., 1994; Zhang et al., 2003]. Summertime anthropogenic NOx emissions over the eastern US have been decreasing since 1999 due to EPA regulations [Frost et al., 2006]; however, the contribution of the anthropogenic emissions to tropospheric NOx remains significantly greater than that from lightning [Hudman et al., 2007]. Consequently, anthropogenic emissions contribute more to the tropospheric O_3 over the US than lightning, except in the convective outflow regions over the North Atlantic [Choi et al., 2008b].

Lightning and anthropogenic emissions affect different parts of the troposphere, the upper and lower troposphere respectively [Zhang et al., 2003; Hudman et al., 2009]. This has important implications on the radiative impact (defined here as a change of net shortwave (<4.0 μm) and longwave (<2200 cm^{-1}) fluxes at the top of the atmosphere) by these two sources. Mid- and upper tropospheric O_3 has a larger radiative forcing efficiency than its lower tropospheric counterpart; for example, an increase in O_3 of 10 Dobson units in the upper troposphere (8-12km) affects the surface temperature six times more than the same increase in the boundary layer (0-2km) [Lacis et al., 1990]. Thus, lightning-derived O_3 can have a stronger atmospheric radiative impact than O_3 from anthropogenic emissions.

A number of studies have analyzed the impact of lightning and anthropogenic NOx emissions (referred to as "LNOx" and "ANOx") on the amounts of tropospheric NOx and O_3 [Zhang et al., 2003; Hudman et al., 2007, 2009; Cooper et al., 2007; Choi et al., 2008a, 2008b]; however, the influence of these sources on the atmospheric radiative impact via O_3 production remains largely unknown. Cooper et al. [2007] calculated positive all-sky adjusted radiative forcing at the tropopause above Huntsville, Alabama of 0.50 Wm^{-2}. The North American Monsoon (NAM) strongly affects both tropospheric dynamics and chemistry over North America [Ridley et al., 1994] and also the vertical structure of O_3 [Zhang et al., 2003; Li et al., 2005; Cooper et al., 2007]. Understanding the impact of changes in tropospheric O_3 at different altitudes on the radiation field is an important step towards improved quantification of radiative forcing in climate studies.

This study examines the influence of the O_3 generated by LNOx and ANOx on the summertime OLR and radiative impact over the southeastern US and the North Atlantic. The Regional chEmical trAnsport Model (REAM) model is used to quantify the tropospheric NO_2 and O_3 enhancements by LNOx and ANOx in conjunction with the satellite data from the Ozone Monitoring Instrument (OMI) and Tropospheric Emission Spectrometer (TES). We perform a control and two sensitivity simulations. The control run includes all NOx emissions. The two sensitivity runs remove either LNOx or ANOx so that the effects of different sources can be extracted by differencing the control run and the corresponding sensitivity run. The simulated OLR is evaluated against observations from the NOAA-16 satellite. The simulation is also used to examine the relative importance of LNOx O_3 over ANOx O_3 contribution to the radiative impact before and during the NAM.

2. Satellite Measurements

The OMI and TES instruments on board the NASA Aura satellite make nadir measurements at 01:45 and 13:45 local time (LT) with the footprints of 13×24 km and 5×8 km, respectively. The OMI tropospheric NO_2 columns [Bucsela et al., 2006] are obtained from the NASA Goddard Earth Sciences Distributed Active Archive Center. Only OMI data with a cloud fraction of <20% are used [Choi et al., 2008b]. The TES version 3 O_3 data are also filtered for quality; only the data that pass the "master" quality flag test [Osterman et al., 2007] are used in the analysis. In order to insure a proper comparison between the REAM results and the TES data, the TES observation operator (referred to as "averaging kernel") is applied to the model profiles [Worden et al., 2007].

The NOAA-16 satellite data are used to locate deep convection and estimate the radiation budget including OLR [Liebman and Smith, 1996]. The satellite crosses the equator twice a day at 01:50 and 13:50 LT (5 minutes after Aura), and the monthly OLR data are available [Liebman and Smith, 1996] from the NOAA/ESRL Physical Sciences Division (http://www.cdc.noaa.gov/cdc/data.inerp_OLR.html).

3. Model Description

3.1 REAM

The set up for the REAM runs, a horizontal resolution of 70 km with 23 vertical layers from the surface to 10 hPa, is the same as in Choi et al. [2005, 2008a, 2008b]. The 2005 summer GEOS-CHEM (version 7.2) global model simulations [Bey et al., 2001] are used to specify the initial and boundary conditions for trace gases. Emission inventories for the combustion and industrial sources as well as the algorithms for soil and biogenic

sources are adopted from GEOS-CHEM, except for NOx from anthropogenic emissions, biomass burning, and lightning. The NOx emissions in the 1999 EPA National Emission Inventory from the Electronic Generation Unit (EGU) and non-EGU point sources in the eastern U.S are reduced by 50% to account for the recent reduction in anthropogenic emissions [Frost et al., 2006; Hudman et al., 2007]. The anthropogenic NOx emissions in Canada and Mexico are from the Sparse Matrix Operator Kernel Emissions (SMOKE) inventory [Kaynak et al., 2008]. Table 1 summarizes the NOx emissions inventory for June-July 2005 used in this study. A lightning production rate of 300 moles per flash of NO is used, which is within the currently accepted range of LNOx emissions of 30-670 NO moles per flash described by Schumann and Huntrieser [2007]. The Synoz (synthetic ozone; ozone released into the stratosphere at the rate of cross-tropopause ozone flux) method proposed by McLinden et al. [2000] is used in GEOS-Chem [Bey et al., 2001] to simulate cross-tropopause transport of O_3.

3.2 Fu-Liou Radiative Transfer Model

Atmospheric radiative transfer including the impact of clouds and aerosols is computed using the Fu-Liou scheme [Fu and Liou, 1993; Gu et al., 2003]. The scheme uses the δ-4-stream approximation for the solar flux and the δ-2/4-stream approximation for the infrared flux to achieve a balance between accuracy and computational efficiency. It accounts for the direct radiative effects of 18 aerosol types and the optical properties of liquid- and ice clouds. Details of the scheme are provided in the two references above. The meteorological and chemical fields for calculating atmospheric radiative transfer are prescribed from MM5 and REAM, respectively, except the aerosol fields (sulfate, nitrate,

ammonium, carbonaceous aerosols, soil dust, and sea salt) that are obtained from a GEOS-CHEM simulation [Park et al., 2004].

4. Results and Discussion

4.1 OMI tropospheric NO_2 column

The simulated NO_2 for June-July 2005 agrees reasonably with OMI retrievals (Figure 1) with relatively high spatial correlations (>0.8). The simulation underestimates the observed values by 11.5% in June but overestimates them by 3.3% in July over North America and the western Atlantic. Model results indicate that the largest regions of lightning-NO_2 generation are over northern Mexico (>2.0×10^{15} molecules cm^{-2}) and the southern US (>1.5×10^{15} molecules cm^{-2}). Compared to the OMI data, the simulation underestimates NO_2 columns over northern California by up to 80%; the simulated NO_2 columns are overestimated by less than 20% in the near coastal region of the Gulf of Mexico and North Atlantic Ocean and by up to 100% over the oceanic regions. These discrepancies between the OMI data and the simulation are generally comparable to the OMI measurement uncertainties, except over the North Atlantic in July. Compared with GOME [Choi et al., 2008a], the NO_2 column from OMI is smaller over the western North Atlantic [Choi et al., 2008b], partially due to the impact of the a priori profiles on the OMI NO_2 retrievals [Choi et al., 2008a]. Over the US, the simulated NO_2 enhancements due to lightning correspond to 38% and 58% of those from anthropogenic emissions in June and July 2005. Note that the lightning-generated NO_2 enhancement increases by 57% in July after the onset of NAM.

4.2 TES tropospheric ozone

The REAM model results can also be used with TES O_3 profiles to investigate the impact of LNOx and ANOx on the vertical structure of tropospheric O_3 (Figure 2). Only the TES retrievals in July 2005 are available. Figure 2 compares the simulated zonal mean tropospheric (surface-225hPa) O_3 mixing ratios against the TES retrievals. The most notable discrepancies between the simulated and observed O_3 fields occur in the lower troposphere between 32°N and 40°N. In part, the discrepancies are associated with the biases in TES retrievals for the lower troposphere in the northern mid-latitudes [Nassar et al., 2008].

The simulation reveals that LNOx and ANOx enhance O_3 in the upper and lower troposphere, respectively (Figure 2). Both the TES retrievals and the simulation show enhanced upper tropospheric O_3 to the south of 40°N where REAM generates LNOx O_3 enhancements of 8-16 ppbv. The O_3 enhancement of 8-30 ppbv in the lower troposphere between 30°N and 45°N from ANOx is much larger than the upper tropospheric lightning-derived O_3 enhancements.

4.3 Outgoing Longwave Radiation (OLR)

The OLR reduction by LNOx and ANOx O_3 is investigated using NOAA-16 observations and model data sampled to match the satellite observations (Figure 3). The OLR is estimated for all-sky conditions (including aerosols and clouds). The simulated monthly OLR agrees with the NOAA-16 measurements with relatively small biases of 4.7% and 5.7% and high correlation coefficients of 0.88 and 0.89 for June and July 2005, respectively. The model overestimates OLR above the Midwest US and the subtropical

western North Atlantic in June 2005. The model overestimates OLR over most of North America except in the western US and northern Canada in July. Both the simulation and observation show that the peak OLR region over western North America migrates northward from June to July following the onset of the NAM.

The influence of LNOx and ANOx O_3 on OLR during June-July is evident over the convective outflow region. The largest NOx production occurs over northwestern Mexico in the simulation (Figure 1); however, the reduction in OLR via O_3 production is largest over the outflow regions (Figure 3) because of the time needed to form O_3 [Choi et al., 2008b]. The LNOx O_3 reduces OLR by up to 0.35 Wm^{-2} over the outflow region in July; three times more than the peak contribution from ANOx (< 0.15 Wm^{-2}). This result is consistent with previous findings that the sensitivity of radiative impact to tropospheric O_3 changes dramatically with increasing altitude, up to the tropopause, due to increase of the temperature contrast between the radiation absorbed and emitted by an ozone increment [Lacis et al., 1990]. Over the region, the reduction of OLR by lightning increases by 64% after the onset of NAM. The number of NO molecules generated by each flash is a highly uncertain term. A calculation using the suggested upper limit estimate of 670 moles/flash [Schumann and Huntrieser, 2007] suggests the impact of lightning NOx on OLR is up to 0.40 Wm^{-2}, which is a 14% increase of the current estimate of 0.35 Wm^{-2}.

4.4 Radiative Effects

Details of the radiative impact of LNOx O_3 in the convective outflow region are examined using the model data. The regions of peak impact due to LNOx and ANOx O_3

during June-July occurs over the eastern US and its coastal regions (Figure 4). These regions of strong sensitivity to lightning are associated with the onset of the NAM. In July, the region of significant convection and lightning appears over Mexico, Texas and southern US where the monsoon is strong. The high rate of lightning flash occurrences over the region in July are also shown in the National Lightning Detection Network data (not shown). Using the in-situ observed lightning flash occurrences, the simulated lightning flashes are constrained from their dependence on MM5-simulated cloud mass flux and convection available potential energy [Choi et al., 2005, 2008b]. The impact of the LNOx on tropospheric O_3 and the corresponding radiative effect are shifted eastward along the outflow regions as was the OLR [Choi et al., 2008b], resulting in large LNOx O_3-induced radiative impact (up to 0.55 Wm^{-2}) over southeastern US and the western North Atlantic Ocean (Figure 4). The shortwave term has similar structure to the OLR, with some increase in the outflow region.

5. Conclusions

The impact of LNOx and ANOx on the OLR and radiative effects in June-July 2005 has been examined using the REAM simulations in conjunction with observations of the tropospheric NO_2 and O_3 data from OMI and TES, and the OLR by NOAA-16. Both OMI data and the simulations show large NO_2 enhancements over northern Mexico, Texas and the southern US after the onset of NAM due to increased lightning activity. The photochemical O_3 production follows in the convective outflow region. The impact of the LNOx O_3 on OLR (up to 0.35 Wm^{-2}) and radiative effects (up to 0.55 Wm^{-2}) in the convective outflow regions are up to three times as large as those from anthropogenic

emissions. The simulated radiative impact in this study compares well with that (0.50 Wm^{-2}) over Huntsville as estimated by Cooper et al. [2007]. Despite the uncertainty in LNOx generation, the relative importance of LNOx O_3 over ANOx O_3 to the OLR and radiative impact over North America is becoming more pronounced due to the large reduction in ANOx in recent years. The radiative impact from lightning-derived upper tropospheric O_3 over the convective outflow regions will be affected by the potential changes in convection due to global warming, a topic for future study.

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Table 1. NOx emissions inventory in the contiguous US for June-July 2005

Source type	June (Tg N)	July (Tg N)
Fossil fuel	0.46[a]	0.47[a]
Lightning	0.19	0.32
Soils	0.05	0.07
Aircraft	0.014	0.014
Biomass	0.005[b]	0.004[b]
Total	0.72	0.88

[a]The EGU and non-EGU NOx emissions from 1999 EPA NEI are reduced by 50% over the 23 eastern US [http://www.epa.gov/ttn/naaqs/ozone/rto/sip/related.html]
[b]Biomass burning NOx emissions are from the Global Fire Emissions Database (GFED) [http://www.geo.vu.nl/users/gwerf/GFED/data]

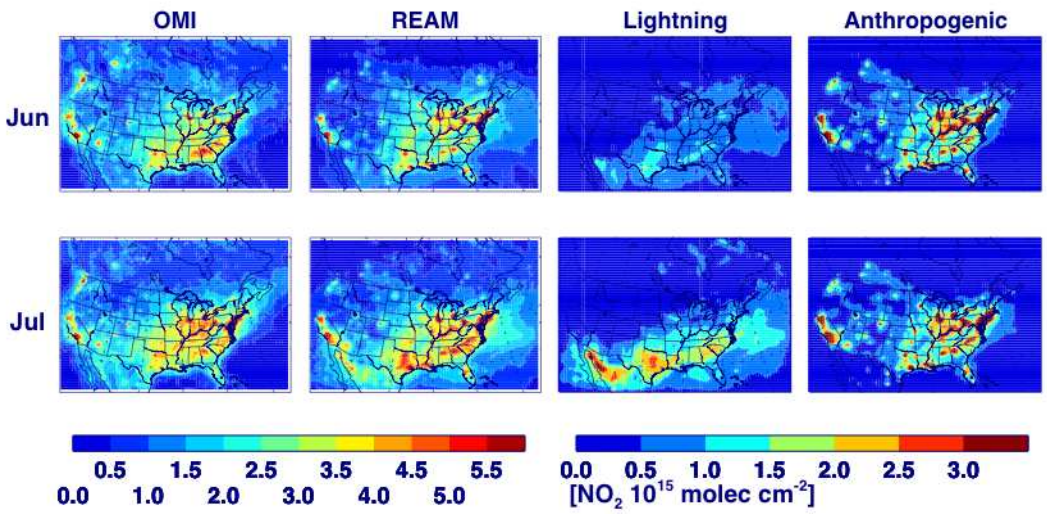

Figure 1. Monthly mean tropospheric NO_2 columns over North America and the western Atlantic for June-July 2005 from OMI satellite measurements (first column), from REAM simulations (second column), produced by LNOx (third column), and produced by ANOx (last column). A different color scale is used for the LNOx and ANOx panels.

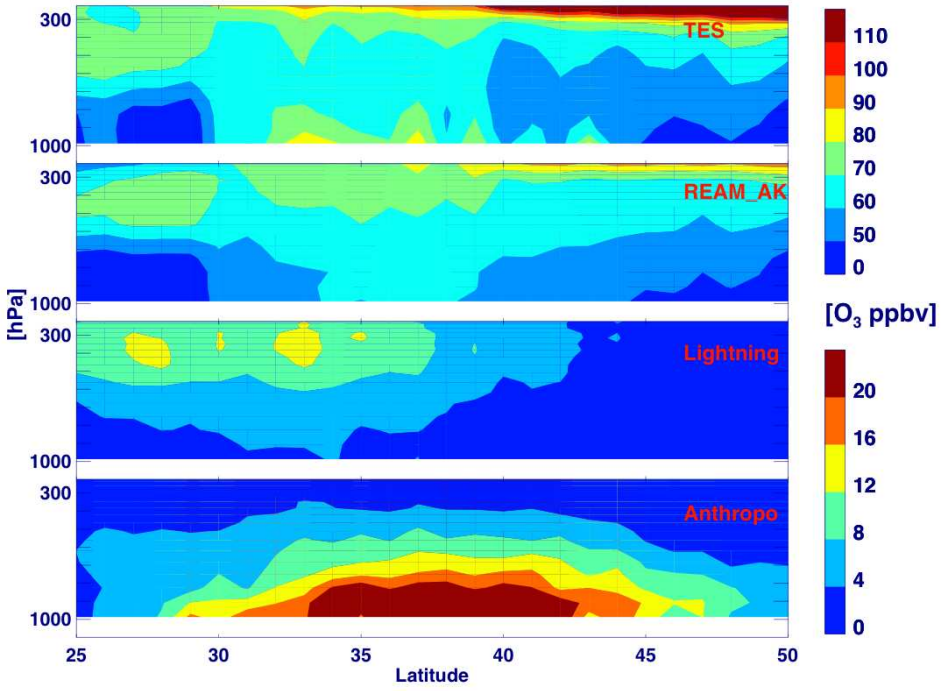

Figure 2. Zonal mean tropospheric O_3 mixing ratios over North America and the western North Atlantic (as depicted in Figure 1) for July 2005 from TES satellite measurements (first panel), REAM with TES averaging kernel applied (second panel), produced by LNOx (third panel), and produced by ANOx (last panel). No averaging kernel is applied to the data in the third and last panels.

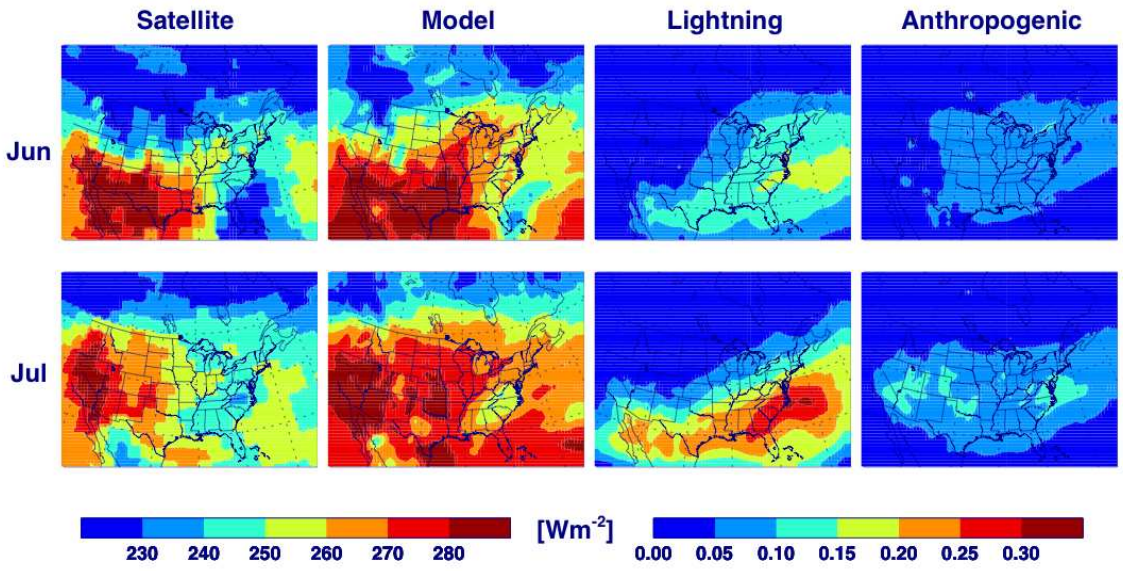

Figure 3. Monthly mean OLR over North America and the western Atlantic for June-July 2005 from NOAA-16 satellite (first column), REAM simulation (second column), and the amounts reduced by LNOx O_3 (third column) and by ANOx O_3 (last column).

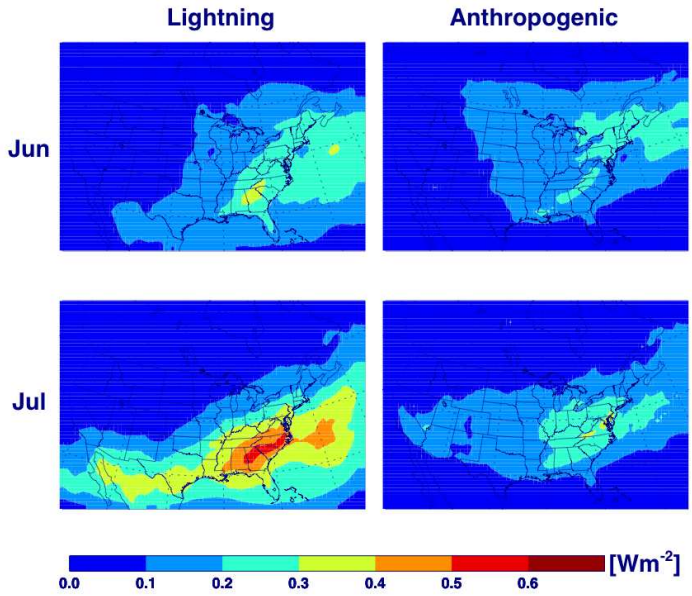

Figure 4. The simulated monthly mean radiative effects (a change of net shortwave and longwave fluxes at the top of the atmosphere) over North America and the western Atlantic for June-July 2005 by LNOx O_3 (left column) and ANOx O_3 (right column).